



APPLICATION 10/681497
APPENDIX

Patent Application of

Stephen G. Bales

for

TITLE: Lignoellulosic, Borate Filled, Thermoplastic Composites

CROSS-REFERENCE TO RELATED APPLICATIONS: 60/427,113-filing 11/18/2002

FEDERALLY SPONSORED RESEARCH: None

SEQUENCE LISTING: None

BACKGROUND:

[0001] This invention relates to lignocellulosic composites, and more particularly, to lignocellulosic, borate filled, thermoplastic composites.

[0002] There is a very high demand for wood products. Although wood is a renewable resource, it takes many years for trees to mature. Consequently, the supply of wood suitable for use in construction is decreasing and there is a need to develop alternatives.

[0003] Lignoelluosic materials, such as wood, sawdust, rice hulls, and the like have long been added to thermoplastic resins such as polyethylene, polypropylene and polyvinyl chlorine (PVC) to achieve a wood-like composite providing reinforcement, reduced coefficient of expansion, and cost reduction. Process methods have been developed to enable blends containing materials having low bulk density (ie. powders) and poor flow characteristics to be fed at commercially acceptable rates. Blends of this type can be extruded through dies of appropriate configuration to produce building product type

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shapes previously made from wood. When these thermoplastic composites were first introduced, the prevailing theory was that the plastic protected the cellulose from fungal attack. However research by Verhey, Laks, and Richer, described in "Laboratory Decay Resistance of Woodfiber/Thermoplastic Composites", Forest Products Journal, September 2001 revealed that lignocellulosic thermoplastics are susceptible to damage from fungal decay. Degradation due to the fungal attack is a problem that threatens the material's structural integrity. In contrast, surface discoloration and spotting has been reported shortly after the introduction of thermoplastic composites. This visual degradation, caused by mold, is a significant problem since major commercial uses of lignocellulosic thermoplastic composites, including decking and fencing, rely on their aesthetic appeal to compete in the marketplace.

[0004] Traditionally, solid wood products are dipped or pressure treated with solutions of fungicides to provide resistance to fungus and mold damage. While this type of treatment is not practicable for a thermoplastic product, it is possible to incorporate a fungicide into the product during its manufacture. This approach provides a constant loading of fungicide throughout the material's thickness, increasing the resistance to leaching of the fungicide from the composite. However it diminishes surface concentration of the fungicide, reducing its effectiveness against surface mold attack.

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Anhydrous borate and zinc borate have been used successfully to provide fungal decay at relatively low levels, typically less than 1.5 percent, in lignocellulosic compounds formed from small fractions of wood bonded with an adhesive binder of phenol-formaldehyde resin as described in US Patent 4,879,083. Zinc borate has also been described in the literature as providing resistance to fungal decay in lignocellulosic filled thermoplastics. Research on zinc borate's use as an anti-fungal additive in lignocellulosic thermoplastics has focused on the minimum loading required to increase resistance to fungal decay, while neglecting to consider or investigate the effect of those higher loading levels required to provide resistance to visual deterioration caused by surface molds.

[0005] Although not used commercially as a fungicide, calcium borate is described in US Patent No 6,368,529 and Patent Application No 20020182431 as providing protection against fungal decay and insects in lignocellulosic compounds formed from small fractions of wood bonded with an adhesive binders of phenol-formaldehyde, phenol-resorcinol-formaldehyde, urea-formaldehyde, and diphenylmethanediisocyanate at preferred levels of 1.5% to 15%. All investigation done on the use of calcium borate as a fungicide has focused on its ability to resist fungal decay in lignocellulosic composites such as particleboard, waferboard, oriented strandboard, and medium density fiberboard that use these thermosetting resins.

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[0006] ~~The use of calcium borate as a fungicide to increase resistance to fungal decay in lignocellulosic thermoplastics is identified in patent application 20030071389 but has not been studied.~~ The use of calcium borate to increase resistance to surface discoloration caused by surface mold has never been identified or studied, either in lignocellulosic composites, which use adhesive binders such as formaldehyde- isocyanate-based resins, and or lignocellulosic thermoplastic composites using resins including polyethylene, polyethylene, and polyvinyl chloride.

[0007] Currently the lignocellulosic thermoplastics industry is faced with two preservation needs: (1) finding an economic method of improving resistance to fungal decay and (2) developing an economic method for improving resistance to the visual damage caused by surface mold.

SUMMARY AND OBJECTIVES OF THE INVENTION

[0008] The present invention, which addresses the above needs, is the incorporation of borates to improve the durability of lignocellulosic thermoplastic products. More specifically it relates to the use of boron-containing fungicides as a preservative to economically increase the resistance of lignocellulosic thermoplastic products to structural decay caused by fungus and to increase the resistance to the visual impairment of the product's surface caused by mold.

[0009] It is an object of the invention is to provide an economic, environmentally safe method of increasing the resistance of a lignocellulosic thermoplastic to fungal decay.

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This is accomplished by the introduction of economic, low toxicity borate materials including calcium borate and boric acid.

[0010] It is a further objective of the invention is to provide an economic, environmentally safe method whereby the lignocellulosic thermoplastic has an increased resistance to surface discoloration and other visual impairments caused by mold. It was discovered this can only be accomplished by ~~increasing~~ testing the borate loading well above the 2 percent level by weight. The invention utilizes the robust nature of the thermoplastic binders to accommodate these increased loadings without creating strength or dimensional problems and resulted in the unexpected discovery that borate loading levels as low as ~~3~~ 1.5 percent produce an improvement, and loadings of about 3 percent produce a significant improvement, in resistance to surface visual impairment caused by mold.

DETAILED DESCRIPTION

[0011] The lignocellulosic thermoplastic composites of this invention are produced by well known procedures that combine molten plastic with lignocellulosic fiber and additional additives such as lubricants, process aids, cross-linking agents, inhibitors, stabilizers, blowing agents, foaming agents and other additives known in the art.

Examples of suitable thermoplastics include polyethylene (PE), high density polyethylene (HDPE), polystyrene (PS), and polyvinyl chloride (PVC) with loadings by weight from 25% to 75%. This process is further described in U.S. Patent No. 5,516,472 (May, 1996). Examples of suitable cellulosic material include wood, ground rice hulls, kenaf, jute, and coconut shells.

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[0012] The methods for manufacturing cellulosic filled thermoplastic are well known and the specific procedure will be dependent on the cellulosic raw material, the plastic, and the type of cellulosic thermoplastic composite desired. However, in general the raw materials are mixed together in a compounding process and the compounded material is then formed into the desired product. Compounding is the feeding and dispersing of fillers and additives, including the fungicide which is in powder form, into the molten polymer using either batch or continuous mixers. The compounded material then is either immediately pressed into the end product or formed into pellets for future processing.

[0013] As used in this invention, the term "boron-containing fungicide" includes calcium borate, zinc borate, and boric acid. The calcium borate which can be used in the method of this invention may be any of the borate compounds containing calcium, boron, and oxygen. ~~The calcium borates include the calcium polytriborates, with a $\text{CaO}:\text{B}_{\text{sub}2}\text{O}_{\text{sub}3}$ ratio of 2:3 and the calcium hexaborates with a $\text{CaO}:\text{B}_{\text{sub}2}\text{O}_{\text{sub}3}$ ratio of 1:3. Calcium hexaborates include nobleite and gowerite. Optionally, calcium-sodium borates and calcium-magnesium borates may be used; examples include ulexite, probertite and hydroboracite.~~ This includes calcium borates that may be synthetically produced or naturally occurring borates including colemanite, ulexite, nobelite, hydroboracite, and gowerite.

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[0014] ~~The preferred boron-containing fungicide for this invention are the calcium polytriborates, which may be synthetically produced or a naturally occurring borate such as colemanite or inyonite.~~

[0015] The exact particle size of the boron-containing fungicide ~~is not critical, but the material~~ must be of a size that can be dispersed uniformly throughout the lignocellulosic thermoplastic composite. Generally a mean particle size as large as ~~150~~ 100 microns and as small as ~~1 micron~~ 3 microns can be used. For best results the mean particle size should be in the range of ~~40~~ 20 microns to 5 microns.

[0016] The amount of boron-containing fungicide incorporated into the lignocellulosic thermoplastic composite will depend on the lignocellulosic content, the longevity desired and the anticipated exposure to moisture. In general, when resistance to decay caused by fungus is required, a range of about 0.2 to ~~5~~ 4 percent by weight of the fungicide is required. The preferred amount is about 0.3 to 2 percent. ~~for lignocellulosic loadings less than 60 percent and about 2 to 4 percent for lignocellulosic loadings greater than 60 percent.~~

[0017] When resistance to visual impairment to the surface caused by mold is required, the amount will be in the range of about 2 1.5 to ~~12~~ 10 percent. The preferred amount is about 3 to 5 percent.

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EXAMPLES

Example 1

Lignocellulosic thermoplastic material was produced using a Brabender Conical Twin Screw Extruder with a counter rotating venting screws and forced through a spring die into test samples 40 cm by 5 cm by 0.2 cm. Prior to extrusion the test samples were blended on a Littleford W-10 mixer as identical mixtures of High Density Polyethylene (HDPE) (>25%), Wood (>40%), Talc (>5%) and Mica (>1%) but with Colemanite loadings of 1, 2, 3, 4, and 5 percent by weight. A sample containing no Colemanite was produced as the control. The Colemanite grade was 47.5 % B.sub.2 O.sub.3 with an average particle size of 7 microns, the HDPE was a BP Solvay virgin reactor flake, and the wood was oak.

The five test samples and the control sample were placed in an outdoor exposure for eighteen (18) months. Visual observations and color recordings using with a Macbeth Color-Eye 7000A spectrophotometer were taken at 6, 12, and 18 months (see Table I). When , after 18 months, the six samples were placed next to one another in ascending order by the level of colemanite content a visible improvement was evident as the level of colemanite increased. The control sample had darkened in appearance considerably more than the colemanite containing samples as can be confirmed by the color data shown in Table 1. Improvement was noted starting with the 1 percent sample, while samples containing 3, 4, and 5 percent colemanite [are] were all relatively similar in color and obtained the best visual appearance of the six sample set.

As the results show, this calcium borate based material can improve resistance to visual impairment caused by surface mold.

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Table 1

Sample	L	a	b	ΔL	Δa	Δb	ΔE
Control							
Initial	64.713	2.266	10.328				
6 month	68.443	0.172	2.879	3.730	-2.095	-7.431	8.574
12 month	64.438	-0.009	1.951	-0.275	-2.276	-8.377	8.685
18 month	63.158	0.007	1.944	-1.555	-2.260	-8.384	8.821
Colemanite 1 %							
Initial	64.968	2.558	10.493				
6 month	69.138	0.181	2.836	4.170	-2.378	-7.657	9.037
12 month	65.673	0.033	2.286	0.705	-2.525	-8.207	8.616
18 month	64.609	0.033	2.406	-0.359	-2.525	-8.087	8.480
Colemanite 2 %							
Initial	65.675	2.382	10.059				
6 month	69.278	0.217	2.903	3.603	-2.165	-7.156	8.299
12 month	67.195	-0.031	2.154	1.519	-2.413	-7.905	8.404
18 month	65.348	0.050	2.469	-0.327	-2.332	-7.590	7.947

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Sample	L	a	b	ΔL	Δa	Δb	ΔE
Colemanite 3 %							
Initial	66.452	2.298	9.921				
6 month	69.884	0.171	2.923	3.392	-2.127	-6.998	8.062
12 month	68.417	-0.021	2.331	1.965	-2.319	-7.590	8.176
18 month	66.678	0.057	2.797	0.226	-2.241	-7.124	7.471
Colemanite 4 %							
Initial	65.957	2.307	9.824				
6 month	68.272	0.195	3.102	2.315	-2.112	-6.722	7.417
12 month	68.208	0.020	2.572	2.251	-2.286	-7.253	7.930
18 month	66.048	0.115	3.075	0.091	-2.192	-6.749	7.097
Colemanite 5 %							
Initial	66.106	2.353	10.087				
6 month	69.661	0.187	3.010	3.554	-2.166	-7.077	8.210
12 month	67.495	0.000	2.514	1.388	-2.352	-7.572	8.050
18 month	66.964	0.066	2.914	0.857	-2.286	-7.173	7.277

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Example 2

Lignocellulosic thermoplastic material was produced using a Brabender Conical Twin Screw Extruder with a counter rotating venting screws and forced through a spring die into test samples 40 cm by 5 cm by 0.2 cm. Prior to extrusion two sets of samples were blended on a Littleford W-10 mixer; set 1 contained Wood (70%), High Density Polyethylene (HDPE) (<30%), Talc (>5%) and Mica (>1%) while set 2 was identical but with the addition of a 2 percent Colemanite loading by weight. The Colemanite grade was 47.5 % B.sub.2 O.sub.3 with an average particle size of 7 microns, the HDPE was a BP Solvay virgin reactor flake, and the wood was oak.

The samples were sanded and trimmed to 3.2 cm by 2 cm by 0.2cm. Labeled samples were supported on plastic mesh in the bottom of beakers to allow water circulation completely around the samples, covered with 250mL of distilled water, and soaked continuously for 14 days at ambient pressure and temperature. The test specimens were then dried at 40°C to dry for 7 days. Then, the test specimens were placed in a 27 °C, 90% humidity environment for 20 days prior to soil block testing.

The soil block test was conducted in accordance with the American Wood-Preservers Association (AWPA) standard procedure E10-91 with the exception that the brown rot samples were placed in jars at the time of inoculation. The white rot fungus *Trametes versicolor* and the brown rot fungus *Gloeophyllum trabeum* were used for the test. Solid wood controls were paper birch and southern yellow pine (SYP) for the white and brown rot tests, respectively as a test of fungal vigor. The following results were obtained:

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Table 2a

SOIL BLOCK TEST RESULTS

White Rot test (*Gloeophyllum trabeum*)

Sample Group	Sample #	Weight Loss%	Average %	Std Deviation %
Untreated				
Birch Control	B-1	67.1		
	B-2	63.9		
	B-3	66.9		
	B-4	66.9		
	B-5	65.1	66.0	1.4
 Sample Set 1				
No preservative	1-1	33.1		
	1-2	39.6		
	1-3	40.4		
	1-4	26.0		
	1-5	37.8	35.4	6.0
 Sample Set 2				
2% Colemanite	2-1	5.1		
	2-2	4.7		
	2-3	2.2		
	2-4	4.8		
	2-5	2.7	3.9	1.4

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Table 2b

Brown Rot test (*Gloeophyllum trabeum*)

Sample Group	Sample #	Weight Loss%	Average %	Std Deviation %
Untreated SYP Control	P-1	62.0		
	P-2	48.6		
	P-3	50.7		
	P-4	43.0		
	P-5	40.8	49.0	8.3
Sample Set 1 No preservative	1-6	38.6		
	1-7	35.2		
	1-8	36.5		
	1-9	38.1		
	1-10	41.7	38.0	2.5
Sample Set 2 2% Colemanite	2-6	11.2		
	2-7	11.8		
	2-8	3.0		
	2-9	9.4		
	2-10	9.6	9.0	3.5

As the above results show, this calcium borate based additive was effective at controlling *Trametes versicolor* and *Gloeophyllum trabeum*. And, as discovered above, at even this relatively low loading the additive would improve resistance to surface discoloration caused by mold.

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CLAIMS

What is claimed is:

1. In the method for forming lignocellulosic thermoplastic composite products containing 25 to 75 percent by weight of the lignocellulosic material such as to increase their resistance to surface visual impairment caused by mold attack, the improvement which comprises incorporating an amount of boron-containing fungicide prior to forming said composite product.
2. The method according to claim 1 in which said amount of fungicide is in the range of from about ~~2~~ 1.5 to about ~~12~~ 10 percent by weight of said composite product.
3. The method according to claim 1 in which said amount of fungicide is in the range of from about 3 to about 5 percent by weight of said composite product.
4. The method according to claim 1 in which said lignocellulosic material is selected from the group consisting of wood, ground rice hulls, kenaf, jute, and coconut shells.
5. The method according to claim 1 in which said thermoplastic material is selected from the group consisting of polyethylene, high-density polyethylene, polystyrene, and polyvinyl chloride.
6. The method according to claim 1 in which said boron-containing fungicide is selected from a group consisting of calcium borate, zinc borate, boric acid, or a mixture thereof.

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~~7. The method according to claim 6 in which said calcium borate is selected from the~~
~~—group consisting of calcium polytriborate, calcium hexaborate, calcium sodium~~
~~—borate, calcium magnesium borate and calcium metaborate.~~

8 7 The method according to claim 6 in which said calcium borate is a naturally
occurring borate.

9 8 The method according to claim ~~8~~ 7 in which said calcium borate is selected from
the group consisting of nobleite, gowerite, hydroboracite, ulexite and colemanite.

~~10~~ 9 The method according to claim 6 in which said calcium borate is a synthetic borate.

~~11 The method according to claim 10 in which said calcium borate is selected from the~~
~~—group consisting of calcium metaborate, calcium polytriborate and calcium hexaborate.~~

~~12 The method according to claim 6 in which said calcium borate is a calcium~~
~~polytriborate having a CaO:B.sub.2 O.sub.3 molar ratio of about 2:3.~~

~~13 The method according to claim 6 in which said calcium borate is a calcium~~
~~hexaborate having a CaO:B.sub.2 O.sub.3 molar ratio of about 1:3.~~

~~14 The method according to claim 1 in which said boron-containing fungicide is selected~~
~~from the group consisting of zinc borate and boric acid.~~

~~15~~ 10 The method according to claim 1 in which said lignocellulosic material is wood.

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- ~~16.~~ 11 In the method for forming lignocellulosic thermoplastic composite products containing 25 to 75 percent by weight of the lignocellulosic material which increase their resistance to fungal attack, the improvement which comprises incorporating an amount of boron-containing fungicide prior to forming said composite product.
- ~~17.~~ 12 The method according to claim ~~16~~ 11 in which said amount of fungicide is in the range of from about 0.2 to about ~~5~~ 4 percent of said composite product.
- ~~18.~~ 13 The method according to claim ~~16~~ 11 in which said amount of fungicide is in the range of from about 0.3 to about 2 percent of said composite product.
- ~~19.~~ ~~The method according to claim 16 in which said amount of fungicide is in the range of from about 2 to 4 percent of said composite product.~~
- ~~20.~~ 14. The method according to claim ~~16~~ 11 in which said lignocellulosic material is selected from the group consisting of wood, ground rice hulls, kenaf, jute, and coconut shells.
- ~~21.~~ 15. The method according to claim ~~16~~ 11 in which said thermoplastic material is selected from the group consisting of polyethylene, high density polyethylene, polystyrene, and polyvinyl chloride.
- ~~22.~~ 16. The method according to claim ~~16~~ 11 in which said boron-containing pesticide is calcium borate, boric acid, or a mixture thereof.

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~~23 The method according to claim 22 in which said calcium borate is selected from the group consisting of calcium polytriborate, calcium hexaborate, calcium metaborate, calcium sodium borate, and calcium manganese borate.~~

~~24~~ 17. The method according to claim 22 16 in which said calcium borate is a naturally occurring borate.

~~25~~ 18. The method according to claim 24 17 in which said calcium borate is selected from the group consisting of nobleite, gowerite, hydroboracite, ulexite and colemanite.

~~26~~ 19 The method according to claim 22 16 in which said calcium borate is a synthetic borate.

~~27 The method according to claim 26 in which said calcium borate is selected from the group consisting of calcium metaborate, calcium polytriborate and calcium hexaborate.~~

~~28 The method according to claim 21 in which said calcium borate is a calcium polytriborate having a $\text{CaO}:\text{B}_2\text{O}_3$ molar ratio of about 2:3.~~

~~29 The method according to claim 21 in which said calcium borate is a calcium hexaborate having a $\text{CaO}:\text{B}_2\text{O}_3$ molar ratio of about 1:3.~~

~~30 The method according to claim 16 in which said boron-containing fungicide is boric acid.~~

~~31~~ 20 The method according to claim 16 11 in which said lignocellulosic material is wood.

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ABSTRACT OF THE DISCLOSURE

The incorporation of borates during the manufacture of lignocellulosic based thermoplastic materials containing 25 to 75 percent by weight of the lignocellulosic material will increase their resistance to surface impairment caused by mold as well as increase their resistance to fungal decay. For resistance to surface impairment, the preferred amount is 3 to 5 percent of zinc borate, calcium borate, or boric acid. When fungal decay resistance is needed the preferred amount is about 0.3 to 2 percent of calcium borate or boric acid.

PROTECTION OF ORIENTED STRANDBOARD WITH BORATE

TREK SEAN
GILLES BRUNETTE
FRANCINE CÔTÉ

ABSTRACT

Experimental panels were prepared to evaluate the effect of borate treatment on the behavior of the treated oriented strandboard (OSB) panels. Polyethylene glycol (PEG) was used as the flowing agent to improve resin fluidity during hot-pressing. The mechanical properties of treated panels containing PEG were found to comply with the CSA 0437 requirements for all properties evaluated. Creep-test results showed no evidence that borate has any adverse effect on the long-term performance of treated panels. The results of leaching tests suggested that panels treated with zinc borate can be used in unprotected environments without any substantial reduction in protection. Decay test results showed that treated panels were not subject to attack by *Gloeophyllum trabeum* nor *Coriolus versicolor*. Weight loss for the treated panels was under 2 percent compared to about 40 to 50 percent for the untreated panels. Formosan termite field tests conducted in Hawaii confirmed that 1 percent of zinc borate provides good protection against termite attack. After 24 months of exposure, panels treated with 1 percent of zinc borate were in a very good condition while the untreated panels were heavily attacked.

Markets for oriented strandboard (OSB) are expanding and the product is being used in more challenging conditions. Like other wood-based materials, OSB panels are susceptible to attack by insects and fungus. There is a need for OSB products that have improved termite and decay resistance, so they can be used under severe conditions. In fact, OSB is being excluded from many applications where customers require improved durability.

The main wood preservatives used in Canada are chromated copper arsenate, pentachlorophenol, and creosote. Attempts to use these traditional preservatives in OSB panels have been made by several researchers (4), but because of the general need for an environmentally friendly treatment, these materials are considered unacceptable for use in a composite board plant.

Borate compounds are considered to have a low environmental impact, and

have even been used as antiseptics (7). Because of its low water solubility, it is very difficult to incorporate zinc borate into solid wood; however, it is quite simple to incorporate it as a powder into composite wood products during the manufacturing process (8). The production of borate-treated panels represents an environmentally sound and highly effective step in the wood and wood products conservation process. The successful utilization of borate as a panel preservative may open many new markets for the OSB industry, especially in countries where the problem of termites

and wood decay are widespread. Furthermore, borate also imparts some fire resistance properties to wood and wood products, which could be an important added benefit (9).

The use of borate as a wood preservative in wood composite panels, however, may cause several problems. The most critical one is related to its adverse effect on the mechanical properties of wood composites bonded with phenol-formaldehyde (PF) resins (6). This difficulty is related to the functional methylol groups (CH_2OH) on resin molecules and their interaction with borate ions (5,7). Such interaction would cause the resin to gel before it is able to develop an effective bond. Severe pressing conditions are required to break such complexes, in order for the resin to be able to flow and cure in a normal manner (5).

The purpose of this study was to develop a process for the successful addition of borate compounds into OSB panels in order to improve the durability of the end products. The design of this experimental work rests on the recognition that the adverse effect of borate on adhesive fluidity can be minimized by the addition of organic flowing agents containing hydroxyl ($-\text{OH}$) groups such as polyethylene glycol (PEG).

The authors are, respectively, Research Scientist, Group Leader, and Technician, Forintek Canada Corp., 319, rue Franquet, Sainte-Foy, Québec, Canada. The authors would like to thank Dr. Peter Laks and his staff at Michigan Technological Univ. for their cooperation, contribution, and support during the termite tests. This paper was received for publication in August 1998. Reprint No. 8854.
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Forest Prod. J. 49(6):47-51.

TABLE 2. — Effect of zinc borate on the fluidity behavior of phenol-formaldehyde resin.

Zinc borate (%) ^a	Diameter of the melted pellet ^b				
	0% PEG ^c	20% PEG ^c	30% PEG ^c	40% PEG ^c	50% PEG ^c
0	49	--	--	--	--
50	44	49	52	64	64 ^d
100	34	42	50	53	58 ^d
200	14	30	35	40 ^d	--

^a Based on resin weight.^b Average of four measurements.^c Based on zinc borate weight.^d Not completely cured.TABLE 1. — Experimental design for the study.^a

Borate content ^b	Resin ^b	Polyethylene glycol
----- (%) -----		
Control (0)	2.5	No
1.0	2.5	No
2.5	2.5	No
5.0	5.0	No
1.0	2.5	40% of borate weight
2.5	2.5	40% of borate weight
5.0	5.0	40% of borate weight

^a Wax content = 1.5 percent based on panel oven-dry weight; panel orientation = random, 26 by 28 by 7/16 inches; target density = 40 pcf including additives, based on oven-dry weight; pressing method = conventional 215°C for 210 seconds.^b Based on panel oven-dry weight.

MATERIALS AND METHODS

PANEL PREPARATION

Experimental panels were prepared in the laboratory using standardized procedures that simulated industrial production. To prepare the panels, commercial aspen strands were dried to a target moisture content of 3 percent. Approximately 20 pounds of dried strands were placed into a drum-type laboratory blender where molten slack wax was sprayed onto the furnish followed by the addition of phenolic resin. After 10 minutes of blending, PEG was sprayed onto the furnish, followed by the addition of zinc borate. The material was allowed to blend for another 10 minutes. Mats were hand-felted onto caul plates prior to hot-pressing, at 215°C for 210 seconds. Other processing parameters are summarized in Table 1.

FUSION DIAMETER TEST

A fusion diameter test was performed to study the flow behavior of the PF resins during hot-pressing. A sample of 0.5 g of resin and mixture was first compressed into a 12.7 by 6.0 mm cylinder (diameter by height). The compressed

pellet was then placed on a hot plate at 150°C between a sheet of aluminium foil and a glass plate. The fluidity of the resin is obtained by measuring the diameter of the melted pellet after 3 minutes of heat under 68 psi of pressure. The sample having higher flow provides larger melted diameter, while smaller melted pellets indicate resin with low flow properties.

PANEL EVALUATION

Mechanical strength and thickness swelling. — Mechanical and physical performance of the panels were evaluated in accordance with the Canadian Standards Association (CSA) standards (3) and American Society for Testing and Materials standards (1).

Leaching resistance. — Leaching tests were performed by immersing panel specimens in flowing water for periods of 6, 24, and 168 hours. The soaking conditions conformed to the CSA water soak test (3). After the immersion period, specimens were oven-dried and milled to wood flour prior to conducting boron analyses.

Decay resistance. — Blocks were exposed to the white- and brown-rot fungi *Gloeophyllum trabeum* and *Coriolus versicolor* in an agar test according to the procedures established by the American Wood-Preservers' Association (2). The weight losses were evaluated based on the oven-dry weight of samples before and after fungal exposure.

Termite resistance. — Termite field tests were performed in collaboration with Michigan Technological University. Samples were exposed to the Formosan subterranean termite at a Michigan Tech test site in Hilo, Hawaii, where test specimens along with untreated wood attractor stakes were placed in a horizontal array on hollow concrete blocks of about 10 cm in thickness. The assembly was

then covered with a wood box to prevent direct contact with rain. This test procedure was designed to simulate wood building components in aboveground applications.

RESULTS AND DISCUSSION

EFFECT OF BORATE ON FLOW BEHAVIOR OF THE RESIN

Table 2 summarizes the effect of zinc borate and PEG on the flow properties of phenol-formaldehyde resin. As expected, the presence of borate reduced the flow properties of the resin. The diameter of the melted pellet decreased with increases in borate content, and this reduction became more and more severe as higher amounts of borate were added. A reduction in diameter of about 75 percent was observed when 66 percent of borate was incorporated in the pellet. This supports the assumption that the borate ions interfere with the functional methylol groups (CH₂OH) of the PF resin. This interaction is known to cause the resin to gel before it is able to develop an effective bond.

An improvement in resin fluidity resulted from the addition of the flowing agent, however, as observed, excessive amounts of PEG also interfered with the curing properties of the resin. The curing time of PF resin increased markedly with the increased in the PEG content.

Resin flow capability was an important parameter controlling the performance of the composite panels. In general, high-flow resins developed larger bonding areas and provided better panel performance as compared to the low-flow resins. This can be applied as a screen test to determine the appropriate amount of borate and PEG to be added to PF resin without interfering with the flowing behavior of the compound.

The diameter of the melted control pellet was about 49 mm. In order to allow the resin to flow, at least in the same manner, the amount of borate and PEG had to be adjusted to have its melted pellet similar or larger than the control. Generally, from the results of different factors and combinations, the viscosity of the melted compound was greatly affected by borate and the maximum level of borate to be mixed with resin was 1 to 1 ratios. Beyond this limit, the compound became very viscous and excessive amounts of PEG were required to improve the flow. This may have created some intermediates, which interfered

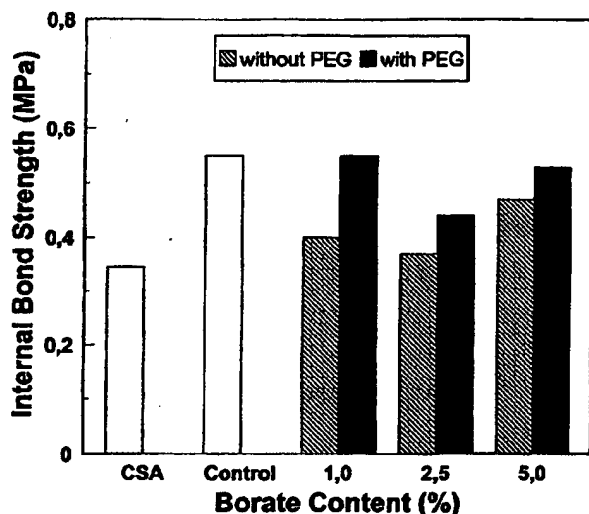


Figure 1. — Effect of zinc borate and PEG on the internal bonding strength of the treated panels.

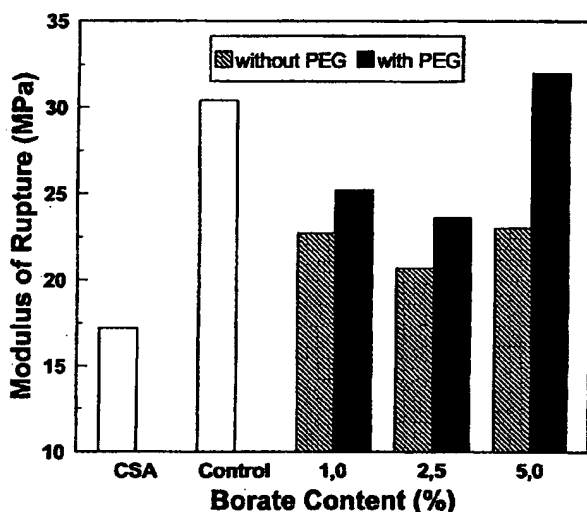


Figure 2. — Effect of zinc borate and PEG on the modulus of rupture of the treated panels.

with the curing properties of the resin. A resin/borate ratio of 100:100 and 40 percent of PEG, based on borate weight, is about 53 mm of diameter and this was considered as an optimum level for this research.

EFFECT OF BORATE ON MECHANICAL PROPERTIES OF THE TREATED PANELS

Figures 1 through 4 show the effect of zinc borate treatment on the change in the mechanical properties of the treated panels. There is a clear indication that al-

though the mechanical properties of the treated panels were somewhat reduced by the borate treatments, this can be overcome by adding PEG into the panel composition. Strength properties of panels treated with 5 percent zinc borate were actually higher than those treated with a lower treatment level. This discrepancy may be explained by the higher resin content used for the panels treated with 5 percent zinc borate. It should be noted that, as shown in Table 1, panels treated with 0, 1, or 2.5 percent borate were

bonded with 2.5 percent resin whereas 5 percent resin was used in the 5 percent borate-treated panels.

Figure 1 shows that the internal bond strength (IB) of treated panels generally decreased with increasing borate content. The addition of PEG into treated panels composition had a positive effect on the adhesive bonds. As expected, the IB strength of the borate-treated panels containing PEG was considerably improved.

The same pattern can be observed with the modulus of rupture (MOR). Figure 2 shows that MOR generally decreased with increasing borate content. Improvement in MOR of the treated panels was observed when PEG was added in the panel composition.

The change in modulus of elasticity (MOE) as a function of borate loading level is presented graphically in Figure 3. There is no clear indication that the MOE of the treated panels was adversely affected by borate treatment. Furthermore, the MOE values of the panels treated with 5 percent zinc borate were higher than the control. Again, higher resin content in the panels may partially or fully account for this observation. Figure 3 also shows that there is some drawback effect of using PEG in the panel composition. MOE values of treated panels containing PEG were generally lower than those with no PEG. This observation was not a surprise and the behavior is mainly related to the plasticizer effect of the PEG compounds.

Figure 4 shows that the durability (aged MOR) of the treated panels was adversely affected by borate treatment. All borate-treated panels without PEG failed to meet the aged MOR requirements. As expected, the addition of PEG produced a dramatic improvement in the aged MOR of the treated panels. All treated panels containing PEG were found to comply with the CSA durability requirements. Clearly, the presence of a flowing agent was necessary to achieve maximum strength for panels containing any level of borate.

Figure 5 demonstrates the change in thickness swelling of the treated panels. It was surprising to discover that dimensional stability of the treated panels was not severely disturbed by borate treatment. No substantial increase in thickness swelling with increasing borate content was noticed. The thickness swelling values were similar for all panels.

The effect of borate on the long-term performance of treated panels is also a concern for the OSB industry. Results of

the creep test presented in Table 3 show that zinc borate has no adverse effect on the long-term properties of OSB panels.

TABLE 3. — Creep properties of zinc borate treated panels.^a

Zinc borate (%)	Cumulative creep	Secondary creep	Irrecoverable creep
0	4.07	0.53	5.08
1.0	3.77	0.54	4.91
2.5	3.14	0.42	4.62
5.0	3.99	0.51	4.82

^a Average of three measurements. load = 10 kg (15% of MOR); humidity = 65% (RH) at 20°C.

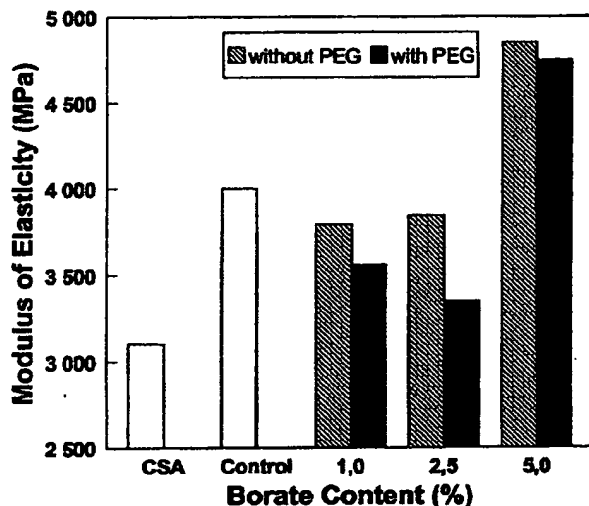


Figure 3. — Effect of zinc borate and PEG on the modulus of elasticity of the treated panels.

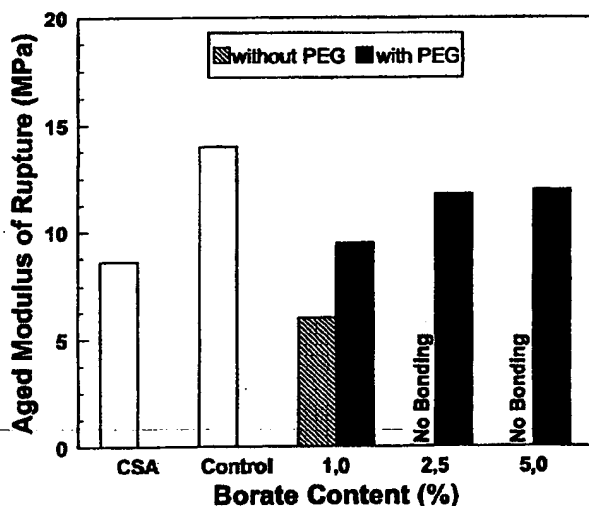


Figure 4. — Effect of zinc borate and PEG on the aged modulus of rupture of the treated panels.

The cumulative, secondary, and irrecoverable creep of the treated panels were generally superior to the control. This indicates that treated panels showed better creep resistance compared to the untreated panel. It is important to note that the CSA test procedure standards require that creep tests be carried out at two constant conditions. The results presented in this study, however, are limited to only one condition and cannot be used as specific values but just as a comparison to the control panels.

LEACHABILITY OF BORATE

Leaching test results are summarized in Table 4. A discrepancy between the target borate content and the measured level in the treated panels was observed. It appears that not all the powder additive was retained on the flakes. However, it was observed that the addition of PEG, a liquid, has a positive effect on the absorption capability of borate and resin onto the surface of the wood flakes. Leaching test results showed that the borate content of treated panels decreased somewhat for short-term leaching, then levelled off after 24 hours of leaching. This suggests that zinc borate treated panels may be used in an unprotected environment without any substantial reduction in protection.

DECAY RESISTANCE

Decay test results are presented in Table 5. Samples from the treated panels were exposed to attack by the white-rot fungus *Gloeophyllum trabeum* or the brown-rot fungus *Coriolus versicolor*. Weight losses for treated panels were under 2 percent compared to 40 to 50 percent for the untreated panels. No fungal growth was observed on the treated panels.

TERMITE RESISTANCE

Formosan termite resistance was evaluated using a 0 to 10 rating scheme. A rating of 10 indicated that the sample was in perfect condition, while a 0 rating signified that the sample was destroyed. Test results presented in Table 6 show that the control panels (no borate) were severely attacked by termites while the treated panels showed little or no damage. After 24 months of exposure, the ratings of control panels were below 5 compared to about 10 for samples containing zinc borate. There is no substantial difference in the ratings between any of the treated samples. This suggests that 1 percent of

zinc borate provides good protection against the Formosan termites.

CONCLUSIONS

Consistent with other studies, borate compounds are very active against a variety of insects and fungi. It is clear from this study that the adverse effect of borate compounds on the mechanical properties of the treated panels can be reduced by simply adding an organic flowing agent containing hydroxyl groups into the panel composition. There is no evidence that borate interferes with the long-term performance of the treated panels.

Zinc borate may partially leach out from the panel. The loading level may need to be adjusted to provide suitable protection against decay and termite attacks.

The successful addition of a less-soluble borate to OSB panels in order to make the products more resistant to decay and insects will be of direct interest to the OSB industry. The production of a treated OSB panel will make the product more suitable in applications where a specific level of termite and decay resistant is required. Furthermore, as a special product, "treated" OSB will be able to command a higher price.

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TABLE 4. — Leachability of zinc borate treated panels.

Zinc borate		Leachability		
Target	In panel	6 hr.	24 hr.	168 hr.
----- (%) -----				
1.0	0.93	0.80	0.75	0.70
2.5	1.92	1.81	1.64	1.44
5.0	3.43	3.0	2.89	2.75

TABLE 5. — Decay resistance of zinc borate treated panels.

Zinc borate	<i>Gloeophyllum trabeum</i>		<i>Coriolus versicolor</i>	
	Weight loss	Fungal growth	Weight loss	Fungal growth
----- (%) -----				
0	> 40	Moderate	> 52	Moderate
1	< 2	none	< 1	none
2.5	< 2	none	< 1	none
5.0	< 2	none	< 1	none

TABLE 6. — Termites attack rating of zinc borate-treated panels.^a

Zinc borate	6-month rating	12-month rating	18-month rating	24-month rating
(%)				
0	9.7	7.1	6.4	4.5
1	10	9.8	9.8	9.8
2.5	10	10	10	10
5	10	10	10	10

^a Average of 10 assessments based on a scale from 0 to 10; 10 indicates sound and 0 indicates completely destroyed.

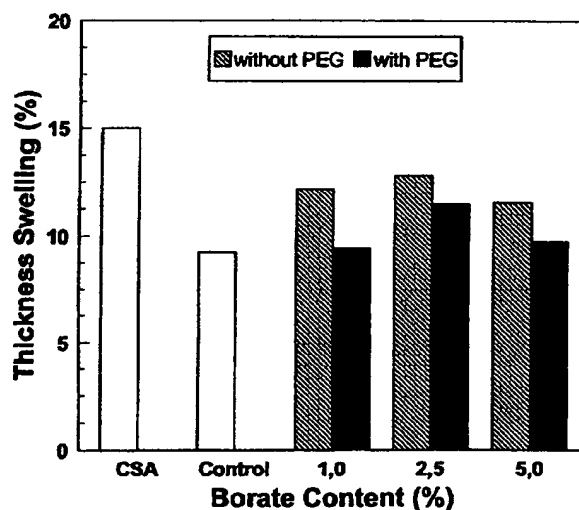


Figure 5. — Effect of zinc borate and PEG on the thickness swelling of the treated panels.